

# Life-cycle assessment of emerging CO<sub>2</sub> mineral carbonation-cured concrete blocks: Comparative analysis of CO<sub>2</sub> reduction potential and optimization of environmental impacts

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# Journal Pre-proof

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Hao Huang, Tao Wang, Ben Kolosz, John Andresen, Susana Garcia, Mengxiang  
Fang, M. Mercedes Maroto-Valer

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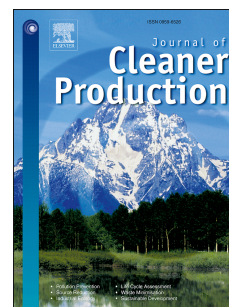
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## Title

Life-cycle assessment of emerging CO<sub>2</sub> mineral carbonation-cured concrete blocks: Comparative analysis of CO<sub>2</sub> reduction potential and optimization of environmental impacts

## Authors

Hao Huang<sup>a,b</sup>, Tao Wang<sup>\*,a</sup>, Ben Kolosz<sup>b</sup>, John Andresen<sup>b</sup>, Susana Garcia<sup>b</sup>, Mengxiang Fang<sup>a</sup>, M. Mercedes Maroto-Valer<sup>\*\*,b</sup>

<sup>a</sup> State Key Laboratory of Clean Energy Utilization, Zhejiang University, China

<sup>b</sup> Research Centre for Carbon Solutions (RCCS), School of Engineering & Physical Sciences, Heriot-Watt University, United Kingdom

\* Corresponding Author: oatgnaw@zju.edu.cn

\*\* Corresponding Author: M.Maroto-Valer@hw.ac.uk

## Abstract

CO<sub>2</sub> mineral carbonation (MC) curing technology provides a promising solution for large-scale CO<sub>2</sub> utilization and construction sectors towards low-carbon and environmentally friendly production of concrete, but studies on the total environmental impacts of this technology are scarce. Accordingly, this paper evaluated the life cycle environmental impacts of seven promising concrete blocks from CO<sub>2</sub> MC curing manufacturing pathways (Ordinary-Portland cement block, MgO-Portland cement block, wollastonite-Portland cement block, limestone-Portland cement block, calcium silicate cement block, slag-Portland cement block and Waste Concrete Aggregate block), offering detailed results of cradle-to-gate life cycle assessment and inventory. Identification of the contributions of subdivided raw materials and manufacturing processes, as well as the energy consumption, transportation, and upstream processes for raw materials was performed. It was shown that 292~454 kg CO<sub>2</sub>-eq global warming potential (GWP) of 1 m<sup>3</sup> CO<sub>2</sub>-cured non-hollow concrete blocks were obtained. By contrast, results indicated the 419kg CO<sub>2</sub>-eq GWP from a base case of conventional (steam-cured, non MC) Ordinary-Portland cement block. Up to 30 % of CO<sub>2</sub> emission avoidance could be achieved when

replacing steam curing by MC curing and adjusting the binder types. From the point of view of materials and manufacturing, the reduced use of Portland cement is a key step for environmental optimization, while reducing the energy consumption for maintaining high-pressure carbonation helps to cut down the cumulative energy demand. Increasing the blending ratio in binary binders and the lightweight redesign also proved to be beneficial solutions for mitigating environmental impacts of CO<sub>2</sub>-cured concrete blocks. Wollastonite-Portland cement block and slag-Portland cement block using natural wollastonite and blast furnace slag in binary binders obtained the most favorably scores in all impact assessment indicators, and thus, are arguably considered as the most sustainable types of concrete blocks.

## Key words

CO<sub>2</sub> mineral carbonation curing; Life cycle assessment; CML2001; Global warming potential; Green concrete

## Highlights

- Life cycle assessments of mineral carbonation cured-concrete blocks were conducted.
- Contributions of subdivided raw materials and manufacturing processes were identified.
- Beneficial solutions for mitigating environmental impacts were determined.
- Replacing steam curing by mineral carbonation curing helps environmental optimization.

## 1. Introduction

Greenhouse gas (GHG) emissions from fossil fuel combustion have attracted significant attention due to increasing political pressure on tackling climate change. Among the numerous carbon dioxide (CO<sub>2</sub>) utilization technologies available, mineral carbonation (MC) of CO<sub>2</sub>, which involves chemical reactions that are analogous to geologic mineral weathering, has been proposed as a promising scalable route (Sanna et al., 2014). CO<sub>2</sub> reacts with alkaline metal minerals to form carbonate products during the MC reaction. To address product market scale limitation of aqueous MC technologies, researchers currently use the MC process of cement for early-stage concrete curing (namely CO<sub>2</sub> MC curing technology (Zhang et al., 2017)), realizing CO<sub>2</sub> fixation and obtaining high-value concrete products. The

1 market scale of concrete products significantly exceeds that of high purity carbonate products  
2 obtained from the aqueous MC technologies, offering a promising CO<sub>2</sub> utilization potential.  
3 The MC curing also differs from the natural carbonation of steel reinforced architecture  
4 products, where CO<sub>2</sub> reacts in a durability deterioration mechanism (Ahmad, 2003).

5 The MC curing technology is capable of replacing energy-intensive steam curing by  
6 accelerating carbonation cementation (Rostami et al., 2012). When CO<sub>2</sub> diffuses through  
7 micropores in the cement matrix, the pore structure is densified with the MC conversion of  
8 calcium silicates and hydrated products to carbonated products. Thus process could replace  
9 the hydration reaction to cure the concrete products. The energy saving potential and raised  
10 level of productivity of this process are well aligned with sustainable strategies of  
11 construction sectors towards low-carbon and environmentally friendly production. However,  
12 data on energy and efficiency of this process are scarce when applying CO<sub>2</sub> as a material  
13 input, as well as total environmental impacts. Several studies on the footprint assessments of  
14 some CO<sub>2</sub>-cured products (MgO-cement paste (Unluer and Al-Tabbaa, 2014), for instance),  
15 have provided knowledge of the potential CO<sub>2</sub> emission reduction about a specific material.  
16 Furthermore, environmental accounting methods such as Life-cycle Assessment (LCA) are  
17 required to establish a comprehensive understanding of manufacturing processes and product  
18 types.

19 LCA has been successfully applied in the construction sectors to evaluate the  
20 environmental impacts of multiple life cycle process pathways (Gursel et al., 2014; Vieira et  
21 al., 2016). For steam-cured or air-cured concrete products, the literature provides results of  
22 impact assessment methods and scenario modelling, measuring various influencing factors  
23 from the aspects of the material (Yang et al., 2015), manufacture (Flower and Sanjayan,  
24 2007), transportation, energy input (Dahmen et al., 2017; Marinković et al., 2017) and so on.  
25 There is, however, very little information available on the LCA of CO<sub>2</sub>-cured concrete  
26 products. Research on hydration (García-Segura et al., 2014; Heede and Belie, 2012) or  
27 alkali-activated concrete products (Dahmen et al., 2017) undoubtedly indicates that carrying  
28 out an LCA study is necessary for the environmental impact comparison involving various  
29 raw materials. It is also beneficial for the identification of key parameters and optimization of

products and approaches in different regions and application scenarios.

To conduct LCA studies of CO<sub>2</sub>-cured concrete, several knowledge gaps regarding the materials and processes should be addressed. Substituting CO<sub>2</sub>-activated binders for carbonation hardening instead of hydration have been widely proposed (Mahoutian and Shao, 2016; Sahu et al., 2013 ; Zhang et al., 2017), allowing less energy use and lower GHG emissions by reducing traditional cement production. Previous studies explored the MC curing feasibility of MgO-Portland cement (Mo et al., 2016), wollastonite-Portland cement (Huang et al., 2019; Wang et al., 2017), limestone-Portland cement (Tu et al., 2016) and slag-Portland cement (Liu et al., 2016), which partially replaced cement by other minerals to form binary binders as the substitute of the tradition Portland cement in concrete. The crushed and recycled waste concrete (Zhan et al., 2016) as coarse aggregate in the CO<sub>2</sub>-cured concrete block has also been investigated from a point of view of mechanical performance. The potential environmental benefit of CO<sub>2</sub>-cured concrete is strongly dependent on the type of raw materials.

The optimizations of mix designs and approaches for CO<sub>2</sub>-cured concrete products also needs to be addressed, especially aiming at improving the environmental impacts. Similar to the widely studied hydration process of hydraulic binders and aggregates, significant differences in reaction mechanisms and structural evolutions exists when considering different CO<sub>2</sub>-activated binders (and reactive aggregates). Researchers and industrial developers are modifying the curing conditions (batch duration, pressure, temperature CO<sub>2</sub> concentration) to fit with different types of concrete blocks, striving to achieve qualified mechanical performance and large CO<sub>2</sub> uptake during MC curing. Wang et al. (Wang et al., 2017) adopted up to 25 bar pressure for the MC curing of wollastonite-Portland cement within a 2-hour reaction. On the other hand, Ashraf et al. (Ashraf et al., 2017) extended the curing to 96 hours in atmospheric pressure and 65°C constant temperature. Since different materials are involved, the corresponding curing conditions and energy input are also required when comparing the environmental impacts of the CO<sub>2</sub>-cured products.

Overall, establishing whether there is sufficient environmental rationality when developing CO<sub>2</sub>-cured concrete blocks is a necessary step before further focusing on the

technological and economic factors. This paper aims to evaluate the environmental impacts and influencing factors of seven promising CO<sub>2</sub>-cured concrete blocks from CO<sub>2</sub> MC curing manufacturing pathways, offering detailed results of LCA and life cycle inventory (LCI). Global warming potential (GWP) (measured in CO<sub>2</sub>-equivalency) and nine other major indicators in the well-known CML-IA (Centre of Environmental Science in Leiden University (Guinée et al., 2002)), as well as two indicators in Cumulative Energy Demand, are selected as impact assessment methods for the environmental comparison of concrete mixes and scenarios. GWP evaluation includes the various contributions subdivided into different raw materials and manufacturing steps. A reference calculation (as a base case) using the baseline mix design of Ordinary Portland Cement Block with steaming curing process compares the GWP difference between MC curing and conventional steam curing. The impact of blending ratios in binary binders and the comparison between standard and lightweight products are also explored, followed by the sensitivity evaluation of the CO<sub>2</sub> uptake, energy consumption, and transportation distance. Finally, optimized mix design and solutions for mitigating environmental impacts of CO<sub>2</sub>-cured concrete blocks are also discussed.

## 2. Methods

The LCA methodology used in this study is based on the widely accepted International Standards Organisation (ISO) 14040 and 14044 (ISO, 2006). A global inventory database, ecoinvent v3.4 (Ecoinvent, 2017) (current version 3.5 as of August 2018) was used to obtain inventory data. As MC curing technology is in its infancy, detailed data of the manufacture processes cannot be derived from established life-cycle inventories. The CML-IA (10 indicators) and the Cumulative Energy Demand (2 indicators) are applied (Braga et al., 2017). An original Excel-based software is used for the calculations for all scenarios. One can repeat the calculations based on the equations in this study.

### 2.1 Goal

The goal of this study was focused on the environmental impacts of different CO<sub>2</sub>-cured

concrete blocks to better understand the potential advantage of MC curing technology and to compare different types of mix designs. This study investigated and identified the contributions of subdivided raw materials and manufacturing processes, as well as the energy consumption, transportation, and upstream processes for all the materials. The LCA results are also used to ascertain opportunities to improve the environmental performance of CO<sub>2</sub>-cured products at different stages of their life cycle and minimize total GHG emissions and other impact indicators per unit product.

## 2.2 Functional unit and scope definition

In accordance with the goal of this study, the calculation of the cradle-to-gate LCA is conducted and the system boundary is presented in Fig. 1. Only the block product is considered, the paste or mortar samples (studied by lab experiments) are not involved because they are not ready-to-use. The use stage of CO<sub>2</sub>-cured concrete blocks (including the transportation of distribution-to-site) is excluded due to the expected similar conditions for all kinds of mixes. End-of-life stage is assumed to be comparable (most wastes are disposed of in landfills) and thus is omitted as well. To quantify the impact assessment indicators of products within the defined system boundary, the inventory values of the indicators are estimated in the form of inputs of resources as well as the outputs of emissions (to air, land and water) associated with the energy use, resource transportation and conversion of resources. In the base scenario, the CO<sub>2</sub> cylinders are considered as the only gas source (existing research mostly use high-pressure 100% CO<sub>2</sub>), and the energy consumption for gas compression and purification are not included based on the adopted processes and boundary setting. The impact of CO<sub>2</sub> capture and transportation would be discussed in the scenario discussion.

The functional unit of this study is 1 m<sup>3</sup> non-hollow concrete block with MC curing, and is set to correspond to the mechanical power of a full-load operation. Seven types of the material mixes (depending on the types of binders or aggregates) for MC curing are included for comparison. Based on the boundary settings, all the mixes for MC curing consist of the binder, fine aggregates, coarse aggregates and water as raw materials. Apart from the Ordinary Portland Cement block (OPC), substitute binary binders with the potential benefit



of environmental or mechanical performance are also considered, including the Wollastonite-Portland Cement Block (WPC), MgO-Portland Cement Block (MPC), Limestone-Portland Cement Block (LPC) and Slag-Portland Cement Block (SPC). The Calcium Silicate Cement Block (CSC) using calcium silicate cement as the binder and Waste Concrete Aggregate block (WCA) using the recycled waste concrete as the coarse aggregate are also involved. Among these types, some of them have been studied or developed by the authors experimentally (OPC, WPC, LPC and SPC) in the lab and the others are referred from the present researches.

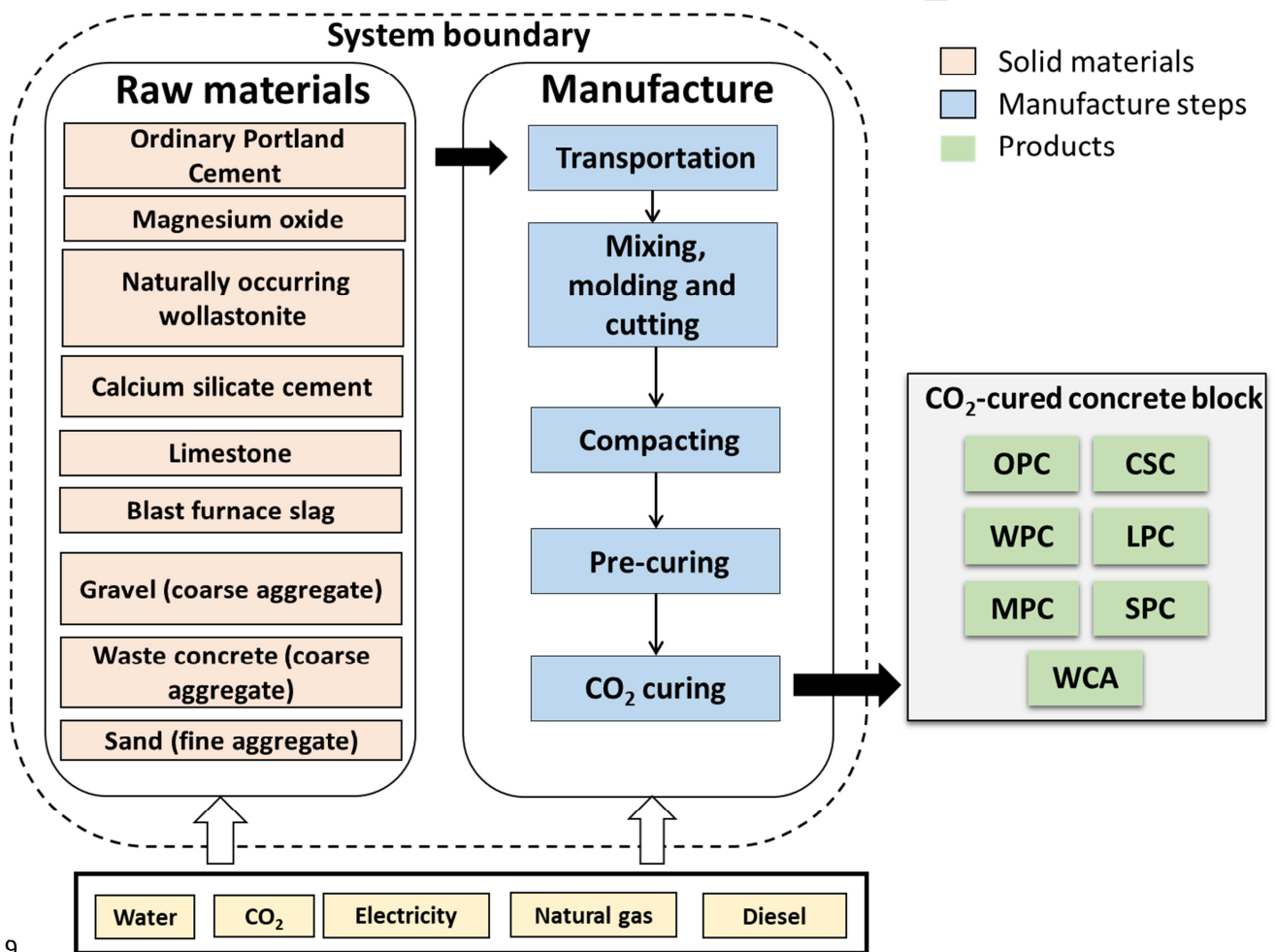


Fig. 1 Life cycle of CO<sub>2</sub>-cured concrete blocks and system boundary

The mix designs of the above CO<sub>2</sub>-cured concrete blocks in the base scenario are presented in Table 1. The design density of each block is 2000 kg/m<sup>3</sup> in the base scenario. The determination of aggregate-to-binder ratio, coarse-to-fine aggregate ratio and water-to-binder

ratio can be referred to in the supplementary data.

The raw materials as input and associated manufacturing processes are also shown in Fig. 1. The manufacturing processes follow a similar sequence before the MC curing stage. The compaction process is necessary due to the binary binders lack of early strength. Therefore at this stage, the processes differ depending on the mix design in the existing literature. A pre-curing process before MC curing is used to control the water-to-binder ratio in order to promote the gaseous diffusion of CO<sub>2</sub>. To establish the life cycle inventory of MC curing, detailed curing conditions (holding pressure, temperature and duration) and the amount of CO<sub>2</sub> required which vary with different types of raw materials are derived from experimental data as listed in Table S1. Only the carbonation conversion ratios of binders are involved. To conduct the MC curing, both the temperature and pressure control are calculated. Heat quantities for reaction temperature are divided into two parts: CO<sub>2</sub> gas and concrete block (as presented in the following section). Energy requirements for pressure control and gas circulation are calculated by considering a general machine operating time (<18.64 KW per m<sup>3</sup>, including light commercial pumps and compressors), while these machines are powered by diesel.

To compare the emission difference between CO<sub>2</sub> MC curing and conventional steam curing, a reference calculation was also conducted by using the baseline mix design of Ordinary Portland Cement Block for steaming curing process. The detailed data for steaming curing was collected from published work (Marceau et al., 2007), which were converted according to the functional unit in this study. The energy input of the steam curing process in manufacture were adjusted to 248.32 MJ energy from natural gas and 97.55 MJ energy from diesel. The emission of using natural gas and diesel was also involved to equivalently compare with previous calculations. The data of steam curing is only applicable to the manufacture of Ordinary Portland Cement block.

1 Table 1 Mix design in base scenario (unit: kg)

2

Type	Portland cement	Coarse aggregate (gravel)	Coarse aggregate (waste concrete)	Fine aggregate (sand)	Wollastonite (naturally occurring)	Calcium silicate cement (calcined)	MgO	Limestone	BFS	Water	CO <sub>2</sub> (not included in the mix mass)	Total mass per m <sup>3</sup>
OPC	370	890	-	592	-	-	-	-	-	148	60.3 (Wang et al., 2017)	2000
WPC	296	890	-	592	74	-	-	-	-	148	68.5 (Huang et al., 2019)	2000
MPC	296	890	-	592	-	-	74	-	-	148	44.4 (Mo et al., 2016)	2000
LPC	296	890	-	592	-	-	-	74	-	148	40.9 (Tu et al., 2016)	2000
SPC	296	890	-	592	-	-	-	-	74	148	42.0 (Liu et al., 2016)	2000
CSC		890		592		370				148	44.4 (Ashraf et al., 2017)	2000
WCA	370		890	592						148	66.6 (Zhan et al., 2016)	2000

3

### 2.3 Description of life-cycle inventory and calculations

Data from ecoinvent v3.4 was used to calculate the life cycle inventory data. Different indicators of input energy for manufacturing processes are carefully considered: electricity is used for compacting, mixing and other plant operations (Dahmen et al., 2017), diesel is used for mineral quarrying, grinding, machine operation in curing and material transportation; natural gas is used for reactor heating (boilers). Both the upstream production and the conversion of energy indicators are involved. The emission data for electricity was obtained from ecoinvent database using an average emission data in China, while the global-average emission data for natural gas and diesel were applied in this study. To calculate the LCI for CO<sub>2</sub> MC curing, the required energy is considered as two parts: (i) the energy input for reactor sealing, pressure control and gas circulation in CO<sub>2</sub> MC curing (CO<sub>2</sub> curing-Pressure, abbreviated as CCP) and (ii) the energy input for reactor and gas heating, maintaining the temperature constant, namely CO<sub>2</sub> curing-Temperature and abbreviated as CCT.

The calculations of different impact indicators could be summarized as:

$$I_i = IR_i + IM_i + IT_i, \quad (1)$$

$$IR_i = \sum_{j=1}^{10} (W_j \cdot IR_{j,i}), \quad (2)$$

$$IM_i = \sum_{k=1}^5 (IM_{k,i}) = IM_{mix,i} + IM_{comp,i} + (IM_{CCT,i} + IM_{CCP,i}) + IM_{TP,i}, \quad (3)$$

$$IM_{GWP} = \left( \sum_{k=1}^5 (IM_{k,GWP}) \right) \cdot C, \quad (4)$$

$$IT_i = \sum_{j=1}^{10} (W_j \cdot D_j \cdot IT_{j,i}), \quad (5)$$

where  $I_i$  is the inventory value of the impact assessment indicator  $i$  ( $i$  is equal to 1-12; 1- global warming potential (GWP), 2- acidification potential, 3- eutrophication potential, 4- human toxic potential, 5- photochemical ozone create potential, 6- abiotic depletion, 7- ozone layer depletion, 8- freshwater aquatic ecotoxicity, 9- marine aquatic ecotoxicity, 10- terrestrial ecotoxicity, 11- cumulative energy demand of non-renewable energy resources (CED-NRe) and 12- cumulative energy demand of renewable energy resources (CED-Re));  $IR_i$ ,  $IM_i$  and  $IT_i$  are the impact assessment data for raw material, manufacturing processed and transportation of material,  $j$  and  $k$  indicate the different raw material types and production steps, respectively (ten types of raw materials include nine solid raw materials and water);  $W_j$  and  $IR_{j,i}$  are the volume weight (kg/m<sup>3</sup>) and inventory value of indicator  $i$  of raw material  $j$ ;  $IM_{mix,i}$ ,  $IM_{comp,i}$ ,  $IM_{CCP,i}$ ,  $IM_{CCT,i}$  and  $IM_{TP,i}$  are the inventory value of indicator  $i$  for mixing,

1 compaction, energy input for CO<sub>2</sub> curing-Pressure (CCP) and energy input for CO<sub>2</sub>  
 2 curing-Temperature (CCT) and material transportation. Especially, for the indicator of GWP,  
 3 the  $IM_{GWP}$  should be calculated as Eq. (4) and  $C$  represents the mass of CO<sub>2</sub> uptake of unit  
 4 product (kg/m<sup>3</sup>); in Eq. (5) the  $D_j$  is the transportation distance (equals to 50 km in the base  
 5 scenario) of raw material  $j$  and  $IT_{j,i}$  is the impact assessment indicator for lorry transportation  
 6 (16-32 ton) (ton<sup>-1</sup>·km<sup>-1</sup>).

$$IM_{CCT,i} = Q_{CCT,i} \cdot (NG_{P,i} + NG_{U,i}) = (Q_{R,i} + Q_{G,i}) \cdot (NG_{P,i} + NG_{U,i}), \quad (6)$$

$$Q_{G,i} = C \cdot \int_{298}^{273+T} C_{p,G} dT = C \cdot \int_{298}^{273+T} (44.14 + 9.04 \cdot 10^{-3} T - 8.54 \cdot 10^{-5} T^2) dT, \quad (7)$$

$$Q_{R,i} = W \cdot \int_{298}^{298+T} C_{p,R} dT, \quad (8)$$

$$IM_{CCP,i} = I_{MO,i} \cdot t = I_{MO,i} \cdot (t_C + t_S), \quad (9)$$

7 where  $Q_{CCT}$  is the total heat needed for temperature maintenance in CO<sub>2</sub> MC curing  
 8 (kJ/m<sup>3</sup>);  $Q_R$  and  $Q_G$  are the heat needed to heat the CO<sub>2</sub> gas and unit concrete block (kJ/m<sup>3</sup>);  
 9  $NG_{P,i}$  and  $NG_{U,i}$  represent the inventory value of indicator  $i$  from the production process and  
 10 utilization (turbine) process of natural gas respectively to supply the corresponding heat; in  
 11 Eq. (7) and (8) the  $C_{p,G}$  and  $C_{p,R}$  are the weight heat capacity of CO<sub>2</sub> and the concrete block,  
 12  $C_{p,R}$  is set to be 0.75 kJ/(kg·°C) (Neville, 2000) and unchanged within range of temperature  
 13 (25-60°C), the total heat from 1 m<sup>3</sup> natural gas is set to be 31.7 MJ;  $T$  is the stable  
 14 temperature for MC curing (K) and  $W$  is the total weight of unit concrete product (kg/m<sup>3</sup>);  
 15  $I_{MO,i}$  is the inventory value of indicator  $i$  of unit general machine operation time (m<sup>-3</sup>·h<sup>-1</sup>) and  $t$   
 16 is the batch duration (h) which consisted of the duration of MC curing ( $t_C$ ) and the silence  
 17 time ( $t_S$ ) between different batches ( $t_S$  which equals 2 hours for atmospheric pressure curing  
 18 and 3 hours for high-pressure curing).

19 Among the raw materials, the production data of Portland cement, magnesium oxide  
 20 (MgO), limestone, gravel, sand, waste concrete and pumice were directly derived from the  
 21 ecoinvent database following the principle of cut-off by classification. The data of Portland  
 22 cement is based on the American data (the data of Portland cement varies considerably based  
 23 on region) while the other materials are based on the global data in the database. The GWP of  
 24 1 kg Portland cement is 0.895 kg CO<sub>2</sub>-eq. The pumice as a lightweight aggregate was only  
 25 used for scenario modelling. Due to lack of data in the ecoinvent, the inventory value of  $IR$   
 26 related to the acquisition of naturally occurring wollastonite (NW), commercial calcium  
 27 silicate cement (CCSC), mainly consisting of synthetic wollastonite, were calculated

separately in the supplementary data. Using the blast furnace slag (BFS) in binary binder also required the energy consumption to reduce particle size by grinding, which was estimated to be 36.7 kWh/ton BFS (from diesel) according to the industrial data. To consider the potential avoided benefit of BFS utilization, this study adopts the No-Allocation principle (BFS is used as merely waste), with no calculation of the mass or economic allocation.

The utilization of waste concrete aggregate in this study was considered as an open-loop process (Yuan et al., 2011): the blocks containing waste concrete aggregate after MC curing would not be recycled as waste concrete. Meanwhile, the avoided burden of waste concrete aggregate was not included, which means that the reuse of waste concrete aggregate produces no extra benefit on the reduced use of energy and resource for the traditional landfill treatment. Last but not least, some assumptions and data for Eq. (3) to (9) regarding the raw materials and manufacturing processes have also been included as supplementary data.

#### 2.4 Scenarios

As mentioned above, the variation in data within the inventories of raw materials and process parameters would significantly affect the outcome of analyzed indicators. For the base scenario, only one density of unit product ( $2000 \text{ kg/m}^3$ ) is adopted, as well as the certain blending ratio of substitute minerals in binary binders (20%). The use of lightweight aggregate may reduce the strength of block, but significantly increases the economics per unit volume of product from the reduction of raw materials and transportation costs. To investigate the possible impacts of product density (associated with the product market and application scenario) and the blending ratio of binary binders, sensitivity analysis would be required. The variations in the amount of  $\text{CO}_2$  uptake and energy for manufacturing processes are also taken into account with the introduction of additional scenarios (3-4). The transportation distance of raw materials, which presents significant impacts on the impact assessment indicators of traditional concrete blocks (Faleschini et al., 2016) strongly affects the availability and use of precast architectural products, and therefore, it is also included. In addition, cost-effective capture and transportation of  $\text{CO}_2$  from industrial processes have proven to be technically challenging in the past and a major limitation of MC processes currently in use. Thus the impact of  $\text{CO}_2$  capture and transportation from industrial processes would be examined by the scenario analysis. The capture of  $\text{CO}_2$  is based on the European data of the production of 1 kg of liquid carbon dioxide out of waste gases from different production processes with the 15-20% monoethanolamine (MEA) solution. The process also

included the purification and liquefaction steps, each using electricity as the energy source. The transportation of gas cylinder is carried out by the lorry (7.5-16 ton, EURO3 emission standard) with the refrigeration machine, the global data was used. The transportation distance of CO<sub>2</sub> is set to be 100 km. The amounts of CO<sub>2</sub> for each type of block are based on the data in Table 1.

A sensitivity analysis is carried out in the form of additional scenarios:

- (1) Scenario 1: The blending ratios of additive in Wollastonite-Portland Cement Block, MgO-Portland Cement Block, Limestone-Portland Cement Block and Slag-Portland Cement Block are decreased to 10% (1A) or increased to 40% (1B);
- (2) Scenario 2: The mix design of seven concrete blocks is reset to fit the standard of the lightweight concrete block (volume density of 1800kg/m<sup>3</sup>, non-hollow), detailed of the lightweight mix designs are shown in Table S3.
- (3) Scenario 3: The energy used in the manufacturing sector is increased (3A)/decreased (3B) by 50%.
- (4) Scenario 4: The CO<sub>2</sub> uptake of the unit product is increased (4A) /decreased (4B) by 20%.
- (5) Scenario 5: The transportation distance of all raw materials is increased by 100% (5A), or only the distance of binder (5B) or aggregate (5C) is increased by 100%.
- (6) Scenario 6: The CO<sub>2</sub> capture and cylinder transportation processes are included.

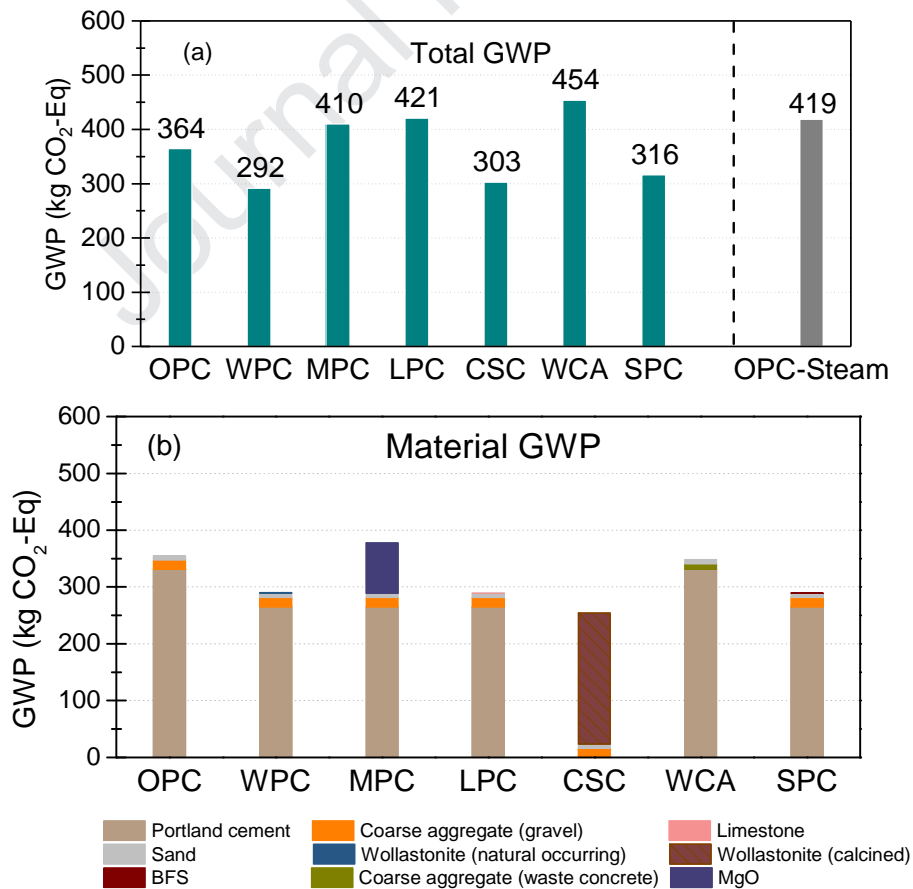
### 3. Results

#### 3.1 Life cycle inventory results and impact assessment

Fig. 2(a) presents the calculated GWP values for all CO<sub>2</sub>-cured concrete blocks and the reference group (OPC concrete block after steam curing). The allocations of GWP for CO<sub>2</sub>-cured concrete blocks by ingredients and manufacturing processes are shown in Fig. 2(b) and Fig. 2(c), respectively. All the results are stated as raw material inputs, manufacture inputs (energy inputs) and impact assessment indicators per m<sup>3</sup> block. Fig. 2(a) shows that Wollastonite-Portland Cement Block presents the lowest GWP of 292 kg CO<sub>2</sub>-eq and Waste Concrete Aggregate Block shows the highest GWP of 454 kg CO<sub>2</sub>-eq. Using the substitute binary binders, the Wollastonite-Portland Cement Block and Slag-Portland Cement Block has lower GWP compared to the Ordinary Portland Cement Block, while the values of MgO-Portland Cement Block and Limestone-Portland Cement Block increase. When compared to the conventional steam curing, the CO<sub>2</sub> MC curing could help to reduce 13.1%

total GWP for the production of Ordinary Portland Cement block, while the Wollastonite-Portland Cement Block using CO<sub>2</sub> MC curing realizes 30.3% total GWP avoidance when compared to Ordinary Portland Cement block using steam curing.

Among various manufacturing processes shown in Fig. 2(c), MC curing (pressure control and gas circulation), concrete mixing (mixing, molding and cutting) and raw material transportation represent the three largest contributions of manufacturing GWP, and the emissions in MC curing mainly depends on its duration, as mentioned above. Only the CO<sub>2</sub> uptakes in the MC curing of Ordinary Portland Cement Block and Wollastonite-Portland Cement Block show the potential to completely offset the carbon emissions in production. It should also be mentioned that the estimation of energy consumption for MC curing process may be overestimated due to the lack of real industry data in this part, especially for blocks which require long curing time. Specifically, in this study when the MC curing is carried out, the functional unit is set to correspond to the mechanical power of a full-load operation, but different machines may have switching and low-load operation periods in actual production, thus the actual energy consumption may be lower than the theoretical value. The adjustment coefficients would need further investigation from the demonstration project.





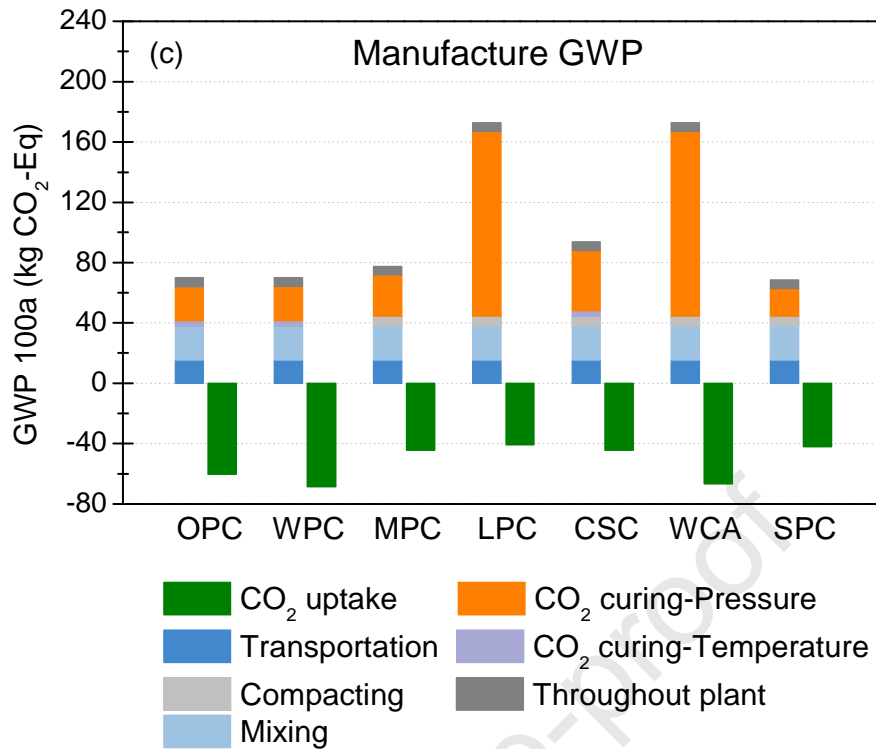
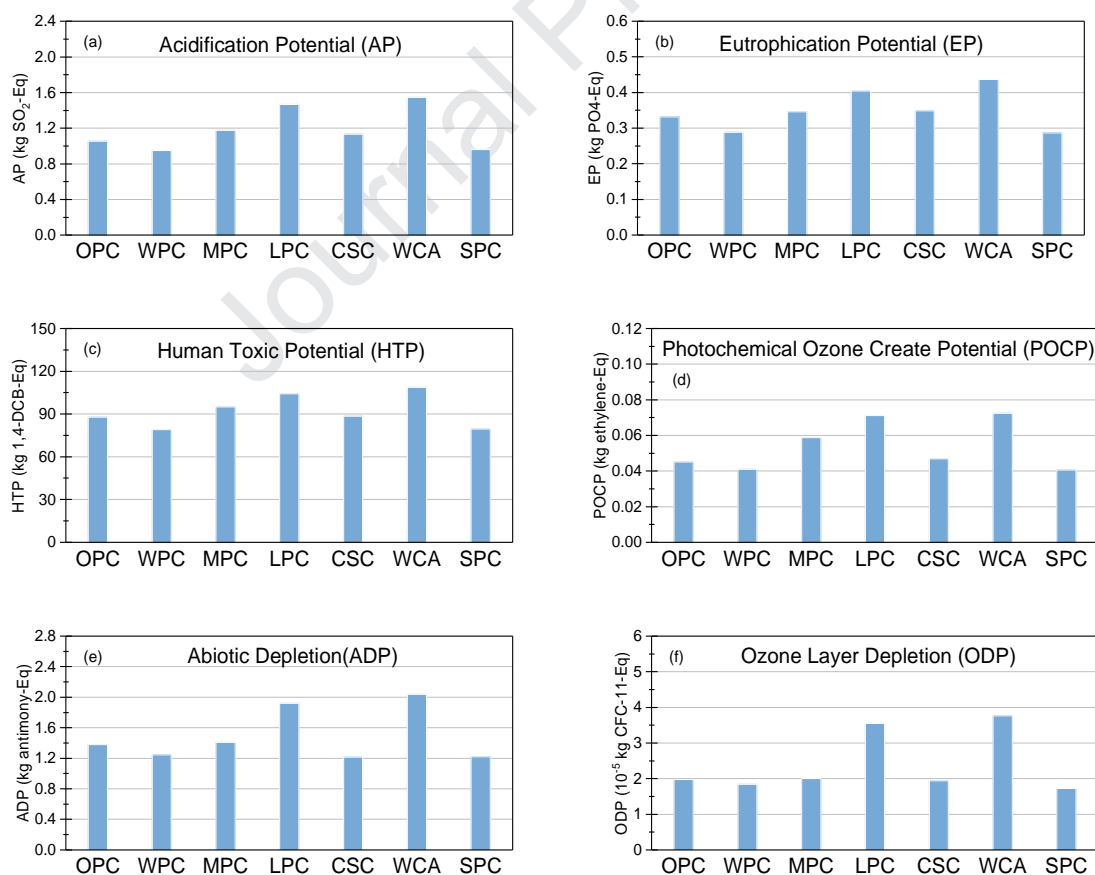


Fig. 2 (a) Total GWP indicator of CO<sub>2</sub>-cured concrete blocks (b) Material GWP distributions  
(c) Manufacture GWP distributions

In a previous laboratory estimation (El-Hassan et al., 2013), 38.65 kWh/(hour·m<sup>3</sup>) was considered for the sample preparation and RH control in the pre-curing step. The authors assumed the MC curing was carried out in a static reactor, ignoring reactor operating energy and other auxiliary equipment. The presented values indicated 96.6 kg CO<sub>2</sub>-eq (0.5 kg CO<sub>2</sub>/kWh (Ecoinvent, 2017)) per m<sup>3</sup> block with 5-hour duration curing, which is higher than the manufacture GWP of Ordinary Portland Cement Block (54.5 kg CO<sub>2</sub> eq) in this study without CO<sub>2</sub> uptake and transportation. Data from the further industrial application of MC curing could also help to improve the energy and emission assessment of manufacturing processes in LCA. It should also be noted that in this study, the energy input for MC curing is strongly associated with the curing duration rather than the curing pressure. This is mainly due to the chosen CO<sub>2</sub> gas source from high-pressure cylinders.

For the base scenario, other impact indicators from CML-IA are shown in Fig. 3. Generally, for different concrete types, similar variations are observed when comparing the 6 assessment indicator results of Acidification Potential, Eutrophication Potential, Human Toxic Potential, Photochemical Ozone Create Potential, Abiotic Depletion and Ozone Layer Depletion. Limestone-Portland Cement Block, and Waste Concrete Aggregate Block score the highest values of all these six impact indicators, and the lowest value is obtained by the Wollastonite-Portland Cement Block and Slag-Portland cement Block. Different diesel used

for curing pressure maintenance should be the main attributor for it. The highest GWP for the MC curing step of Limestone-Portland Cement Block and Waste Concrete Aggregate Block have been mentioned previously, which are associated with their highest diesel consumption. Despite various materials used as binders or aggregates for CO<sub>2</sub> MC curing, the energy input for reactor sealing, pressure control and gas circulation in MC curing would remarkably affect the total impact indicators of Acidification Potential, Eutrophication Potential, Human Toxic Potential, Photochemical Ozone Create Potential, Abiotic Depletion and Ozone Layer Depletion. Meanwhile, using naturally occurring wollastonite and blast furnace slag in binary binders helps to weaken the above six impact indicators compared to Ordinary Portland Cement Block using pure cement (with identical 5-hour batch duration). For MgO-Portland Cement Block, used MgO and prolonged MC curing (6-hour MC curing) present negative impacts, leading to higher impact assessment indicators than those of Ordinary Portland Cement Block. Comparatively, although CSC required 8.8 hours as batch duration, it still presents slight superiority over the Ordinary Portland Cement Block for above six indicators.



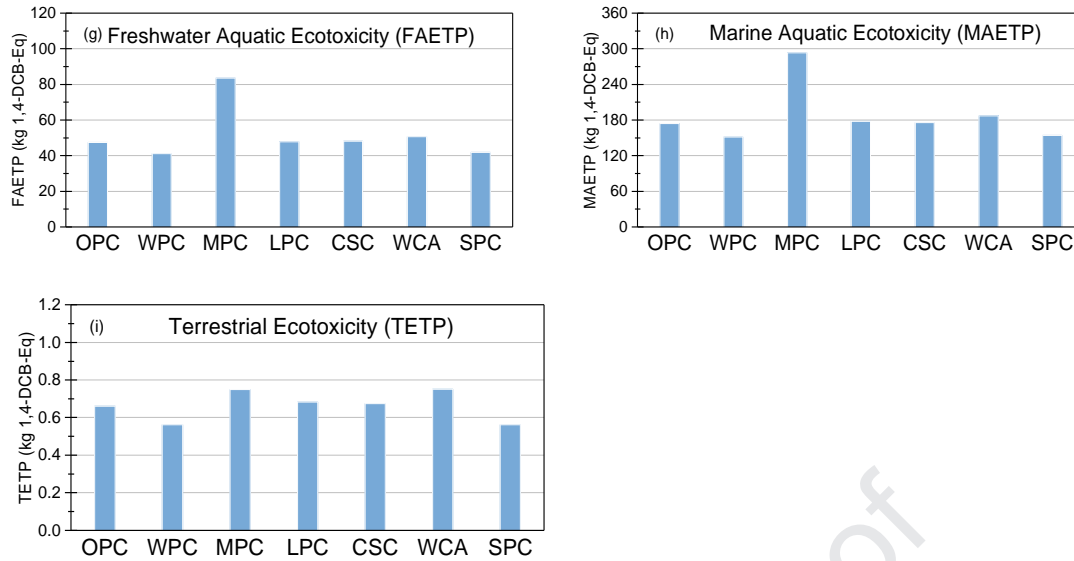


Fig. 3 Results of impact indicators for different CO<sub>2</sub>-cured concrete blocks

Considering the Freshwater Aquatic Ecotoxicity and Marine Aquatic Ecotoxicity in Fig. 3(g) and Fig. 3(h), the major contribution for the highest value of MgO-Portland Cement Block should be given by the production of MgO. Despite the similar manufacture GWP in MC curing when comparing the Ordinary Portland Cement Block and MgO-Portland Cement Block in Fig. 2(c), using MgO instead of Portland cement does not present significant environmental impact benefits. Lastly, for impact indicators of Terrestrial Ecotoxicity, the MgO-Portland Cement Block and Waste Concrete Aggregate Block reached the highest values, which should be associated with the use of MgO and waste concrete aggregate. Previous study (Ruan and Unluer, 2016) has indicated that the coal used for the production of MgO might be the main reason for the high value of Ecotoxicity. When 30% of the coal was replaced by gasoline, the value of Ecotoxicity for the production of MgO would reduce over 90%.

The results of the Cumulative Energy Demand of CO<sub>2</sub>-cured concrete blocks are shown in Fig. 4. The required amounts of primary renewable energy are significantly less than that of non-renewable energy. Both the CED-NRe values of Limestone-Portland Cement Block and Waste Concrete Aggregate Block exceed 4000 MJ-eq, mostly because of the diesel used to maintain the CO<sub>2</sub> pressure during curing. Among these, Slag-Portland Cement Block requires the least non-renewable energy for the unit volume of concrete block, followed by Calcium Silicate Cement Block and Wollastonite-Portland Cement Block.

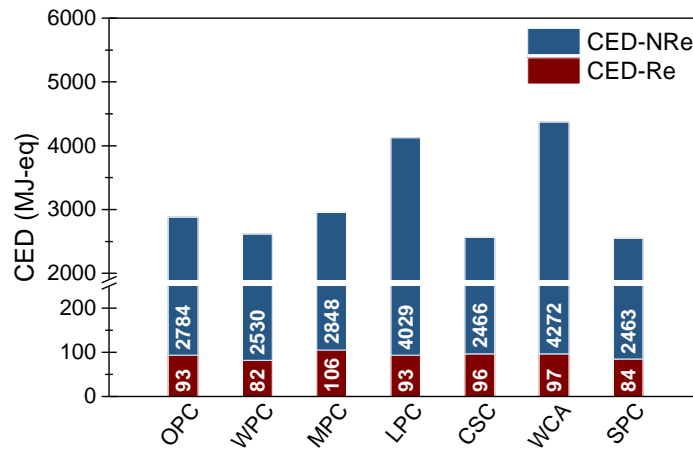


Fig. 4 Cumulative energy demand for different CO<sub>2</sub>-cured concrete blocks

### 3.2 Scenario modelling

Table 2 presents the variations of impact indicators (compared to the base case scenario) for the five scenarios analysed in this study.

#### 3.2.1 Impact of blending ratio of binary binder

The use of the alternative binary binder, especially the replacement ratio of Portland cement in the binary binder, would significantly affect the impact indicators of raw materials and unit product. Thus, previous researchers have worked on replacing as much cement as possible by supplementing cement-based materials to optimize the environmental benefit of the product and avoid much loss of mechanical strength (Yang et al., 2015). In this study, four binary binders have been analysed. For the base case scenario, 20% of the blending ratio (alternative mineral substituting Portland cement) is used, because the binary binders (20%) offer similar or even higher compressive strength as pure cement after MC curing (Mo et al., 2015; Wang et al., 2017). For Scenario-1, the replacements of alternative materials for Portland cement are adjusted to between 10% and 40% (i.e., Wollastonite-Portland Cement Block-10% Wollastonite (WPC-10) or Wollastonite-Portland Cement Block-40% Wollastonite (WPC-40) to explore the potential changes of GWP and cumulative energy demand (CED, sum of CED-NRe and CED-Re). The amount of CO<sub>2</sub> uptake for specific binder was derived from literature (as presented in Table 1), while the curing conditions remained unchanged. Meanwhile, the weight ratio of CO<sub>2</sub> uptake was assumed constant when changing the studied products from paste/mortar to the concrete block.

Fig. 5 shows that GWP of the Wollastonite-Portland Cement Block, Limestone-Portland Cement Block and Slag-Portland cement block decrease as expected with the increment of

blending ratio from 10% to 40%. Among these, GWP of Limestone-Portland Cement Block-40%Limestone (LPC-40) reaches 365 kg CO<sub>2</sub>-eq, close to the GWP of Ordinary Portland Cement Block (364 kg CO<sub>2</sub>-eq), while the GWP of WPC-40 and Slag-Portland cement block-40%Slag (SPC-40) decrease GWP of Ordinary Portland Cement Block by 37.1% and 29.6% respectively. Meanwhile it was observed that WPC-40 produces much lower GHG than that of Calcium Silicate Cement Block (303 kg CO<sub>2</sub>-eq), despite the considerable amount of Portland cement used in mix type of WPC-40. Thus, when using NW, BFS and limestone in the binary binder for MC curing, the increment of blending ratio is beneficial to the reduction of total GWP, as well as other impact indicators (as shown in Table 2). As an exception, the GWP of MgO-Portland Cement Block slightly increases from MgO-Portland Cement Block-10%MgO (MPC-10) to MgO-Portland Cement Block-40%MgO (MPC-40), mainly because of the higher GWP for MgO production (in ecoinvent database (Ecoinvent, 2017)) compared to traditional Portland cement. This is similar to the results previously reported (Mo et al., 2016). As suggested in Section 2.1, replacing Portland cement by MgO fails to achieve a superior performance of impact indicators, and hence, the effort of increasing the blending ratio in MgO-Portland Cement Block would be a negative result. Further process optimization of MgO production and MC curing may be essential. Similarly, the CED values of Wollastonite-Portland Cement Block, Limestone-Portland Cement Block and Slag-Portland cement block including CED-Nre and CED-Re also decrease with the increasing blending ratio, while the values for MgO-Portland Cement Block are almost identical.

1 Table 2 Changes of GWP and CED in scenarios compared to the base case scenario  
2

Type		Scenario-1		Scenario-2		Scenario-3		Scenario-4		Scenario-5	
		10% blending ratio (1A)	40% blending ratio (1B)	Lightweight (LW)	CO <sub>2</sub> uptake +20% (3A)	CO <sub>2</sub> uptake -20% (3B)	Energy +50% (4A)	Energy -50% (4B)	Distance +100% (5A)	Binder distance +100% (5B)	Aggregate distance +100% (5C)
Ordinary Portland Cement Block	GWP			-33.80%	-3.31%	-3.31%	7.48%	-7.48%	4.21%	0.84%	3.37%
	CED			-27.02%			12.79%	-12.79%			
Wollastonite-Portland Cement Block	GWP	9.90%	-21.54%	-32.64%	-4.69%	-4.69%	9.35%	-9.35%	5.26%	1.05%	4.21%
	CED	2.92%	-10.16%	-27.08%			14.10%	-14.10%			
MgO-Portland Cement Block	GWP	-4.51%	7.22%	-33.83%	-2.16%	-2.16%	7.57%	-7.57%	3.74%	0.75%	2.99%
	CED	0.21%	-0.42%	-25.92%			13.97%	-13.97%			
Limestone-Portland Cement Block	GWP	6.63%	-13.26%	-24.55%	-1.94%	-1.94%	18.70%	18.70%	3.65%	0.73%	2.92%
	CED	3.68%	-7.36%	-15.62%			27.71%	-27.71%			
Slag-Portland cement Block	GWP	8.54%	-18.96%	-34.70%	-2.65%	-2.65%	8.39%	-8.39%	4.85%	0.97%	3.89%
	CED	5.25%	-11.21%	-28.63%			13.46%	-13.46%			
Calcium Silicate Cement Block	GWP			-28.87%	-2.93%	-2.93%	12.94%	12.94%	5.07%	1.01%	4.06%
	CED			-19.62%			21.02%	-21.02%			

Waste Concrete Aggregate Block	GWP	-24.58%	-2.93%	-2.93%	17.33%	17.33%	3.38%	0.68%	2.71%
	CED	-15.13%			26.15%	-26.15%			

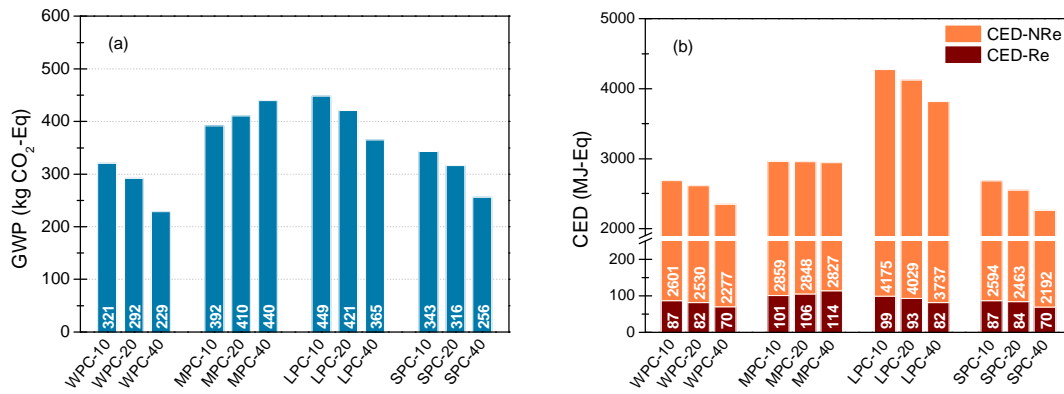


Fig. 5 Sensitivity analysis on blending ratios of binary binders in terms of (a) GWP (b) CED

### 3.2.2 Impact of changing the mix design towards the lightweight block

As a promising alternative to the normal-weight concrete block in the construction sector and off-shore structures, lightweight concrete blocks possess several advantages, including acoustic and thermal insulations, light self-weight which allows easier transportation and higher seismic resistance capacity in building structures (Mehta and Monteiro, 1993). To enable the comparison based on a uniform calculation, the non-hollow structure for one cubic meter concrete block is used. In Scenario-2, the mix designs of lightweight CO<sub>2</sub>-cured blocks (for example, Ordinary Portland Cement Block-Lightweight) are presented in Table S3. In the lightweight design, gravels as coarse aggregate (in the base scenario) are replaced by pumices (except for the Waste Concrete Aggregate Block). The aggregate-to-binder ratio and coarse-to-fine aggregate ratio are redesigned as 7:1 and 1:1 respectively, reducing 10% volume density (from 2000 kg/m<sup>3</sup> to 1800 kg/m<sup>3</sup> for all seven types of blocks).

Overall, after the variation of lightweight design, significant reductions of GWP and CED for all CO<sub>2</sub>-cured blocks are observed (as shown in Fig. 6), and all the reducing ratios exceed 10%, as presented in Table 2. Fig. 6(a) shows that the MgO-Portland Cement Block-Lightweight and Ordinary Portland Cement Block-Lightweight reduce the highest amount of GWP (-139 and -123 kg CO<sub>2</sub>-eq) compared to those in the base case scenario. Slag-Portland cement Block-Lightweight and Wollastonite-Portland Cement Block-Lightweight are the most preponderant types considering GWP reductions (up to -34.7%) and the lowest GWP values after weight reduction. It could be speculated that on the one hand, the Wollastonite-Portland Cement Block and Slag-Portland Cement Block, with low emissions from manufacture processes (in terms of both value and proportion, as supported by Fig. 2), could realize better CO<sub>2</sub> emission controls by improving the material design of the unit product. For the CED indicator, the Waste Concrete Aggregate



Block-Lightweight and Limestone-Portland Cement Block-Lightweight present the smallest values of CED reduction after material changes, mostly because of their high energy-consumption in manufacturing processes as discussed in Section 3.1. Thus, effective optimization of CED should be focused on the manufacturing processes. In terms of other block types, Slag-Portland Cement Block-Lightweight represents the lowest CED of 1765 MJ-eq, while Ordinary Portland Cement Block-Lightweight reaches similar CED as Calcium Silicate Cement Block-Lightweight. This means that the CED of Ordinary Portland Cement Block is easier to improve by optimizing the mix design as compared with CSC (using non-hydraulic binder).

GWP of traditional lightweight concrete blocks containing pumice (air-curing) is found to be 0.2514 kg CO<sub>2</sub>-eq/kg in ecoinvent v3.4 (Ecoinvent, 2017), which is 452 kg CO<sub>2</sub>-eq if applying the density of 1800 kg/m<sup>3</sup> in Scenario-2. In comparison, 1 m<sup>3</sup> CO<sub>2</sub>-cured lightweight concrete block generally produces 196-342 CO<sub>2</sub>-eq as presented in Fig. 6, indicating the avoidance benefit within 24.3-56.6%.

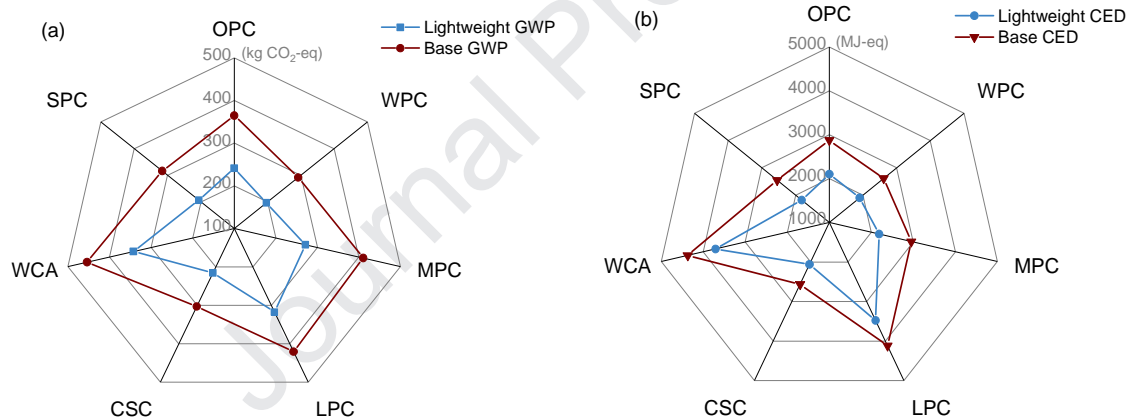


Fig. 6 Comparison of CO<sub>2</sub>-cured concrete blocks in scenario-2 (lightweight products) and base scenario in terms of (a) GWP (b) CED

### 3.2.3 Impact of CO<sub>2</sub> uptake and energy consumption in manufacturing processes

Scenario-3 and Scenario-4 present the influence of altering CO<sub>2</sub> uptake and energy consumption in the manufacturing processes. CO<sub>2</sub> uptake is strongly associated with raw materials and curing conditions, as researchers found that prolonging the duration of MC curing and elevating the CO<sub>2</sub> pressure helps to increase the CO<sub>2</sub> uptake of a specific product. But the kinetic study also showed that after a certain duration (2 hour for OPC (Wang et al., 2017)), the impact of duration time on CO<sub>2</sub> uptake would be neglected. Considering the limitation of theoretical conversion rates (calculated by the amount of alkali metal oxide, ~50

wt. % for Portland cement (Wang et al., 2017)) and carbonation kinetics (reaction rate reduce to near zero after reaching the plateau (Wang et al., 2017)), only 10-20% variations on the CO<sub>2</sub> uptake are investigated in this section. Fig. 7 exhibits the variation of GWP without changing other parameters (material or manufacture). The Wollastonite-Portland Cement Block and Ordinary Portland Cement Block correspond to the highest GWP decline ratios, -4.7% and -3.3% when increasing 20% CO<sub>2</sub> uptake. A 20% CO<sub>2</sub> increase in uptake only causes 1.9-2.2% difference in overall GWP for MgO-Portland Cement Block and Limestone-Portland Cement Block as shown in Table 2. As a whole the impact of CO<sub>2</sub> uptake on overall GWP of CO<sub>2</sub>-cured blocks is limited.

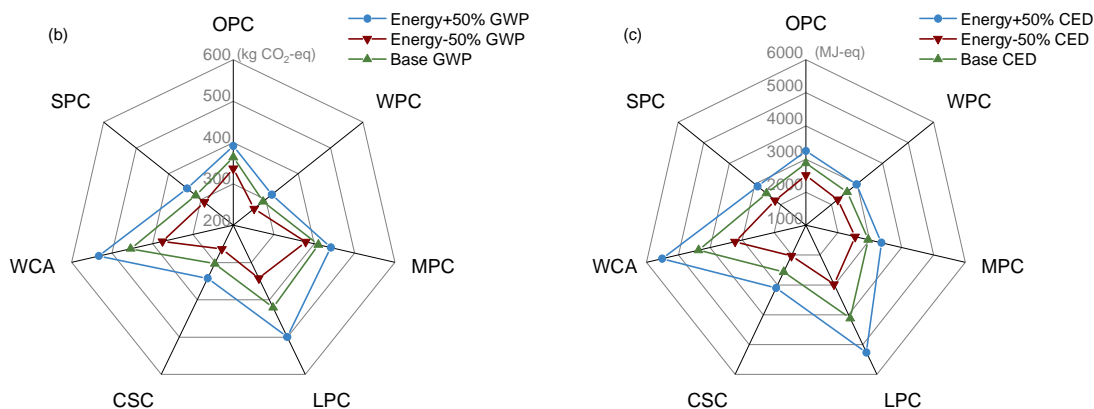
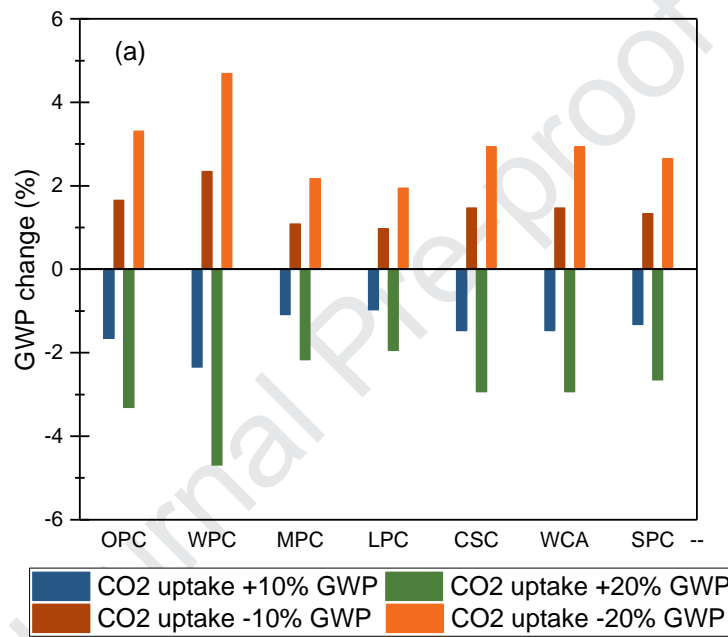


Fig. 7 Comparison of CO<sub>2</sub>-cured concrete blocks when changing (a) CO<sub>2</sub> uptake and (b)(c) energy consumption

In Fig. 7(b)-(c) the sensitivities of energy consumption in manufacturing processes are

presented. The  $\pm 50\%$  variation range set for energy consumption is mainly based on the consideration of curing duration. For example, increasing the duration time from 5 hours to 7 hours would increase over 30% total energy consumption according to the experimental observations. Thus the variation range was increased to 50% in the Scenario-3. Increasing 50% energy consumption would not affect the ranking of GWP for different types of blocks, while decreasing half the energy required for manufacturing results in Calcium Silicate Cement Block possessing the lowest GHG emissions and energy consumption in the study (263.7 kg CO<sub>2</sub>-eq and 2023 MJ-eq vs. 264.4 kg CO<sub>2</sub>-eq and 2244 MJ-eq from Wollastonite-Portland Cement Block). This means that there is a higher energy dependency when using calcium silicate cement rather than wollastonite-Portland cement. It is also clear that MgO-Portland Cement Block produces the largest amount of GHG emissions (379.1 kg CO<sub>2</sub>-eq) with 50% energy saved, implying its limited GWP benefit from process improvement compared to others. Among these, the Ordinary Portland Cement Block shows the least energy dependency of GWP and CED as shown in Table 2 (7.5% GWP increment and 12.8% CED increment for 50% extra energy consumption). The Wollastonite-Portland Cement Block and Slag-Portland Cement Block show similar low values of CED based on their shortest curing duration time, while the large amounts of energy use for MC curing make Limestone-Portland Cement Block and Waste Concrete Aggregate Block the two largest energy users.

### 3.2.4 Impact of transportation distance of raw materials

Scenario-5 (increasing the transportation distance from 50 km in the base case scenario to 100 km) was designed to investigate the impact of transportation distance for the environmental impacts of CO<sub>2</sub>-cured blocks. As presented in Fig. 8 and Table 2, when comparing Scenario-5A and base case scenario the increment of transportation distance results in insignificant changes (mostly within 5%) on GWP for all types of blocks. Partially increasing the transportation distance of binder materials (5B) and aggregate materials (5C) result in similar trends of GWP change, while the increment in Scenario-5B shows a negligible impact when compared to Scenario-5C. Accordingly, the transportation distance of aggregate materials should be given the priority to that of binder materials.

### 3.2.5 Impact of CO<sub>2</sub> capture and cylinder transportation

Fig. 9 (a) shows that the increment of GWP for 1 m<sup>3</sup> CO<sub>2</sub>-cured concrete block is mostly

within the range of 30-50 kg CO<sub>2</sub>-eq with the inclusion of CO<sub>2</sub> capture and transportation processes, which is related to the amount of CO<sub>2</sub> used in curing. And changing the transportation distance of cylinders from 100 to 200km would not cause significant increment. When adding this part of GWP to the total GWP, it can see from Figure 9 (b) that the total GWP of CO<sub>2</sub>-Cured block increases. Among them, the total GWP of OPC is still lower than that of steam-cured OPC (compared with Fig. 2) when considering the impact of CO<sub>2</sub> capture and transportation. But the environmental benefit of MC curing has been significantly weakened. Low GWP values are obtained by WPC, CSC and SPC in Scenario-6, similar to that of base scenario. It is thus clear that the cost-effective CO<sub>2</sub> capture and transportation from industrial processes are essential if these processes are not regarded as external steps out of the boundary of CO<sub>2</sub> utilization.

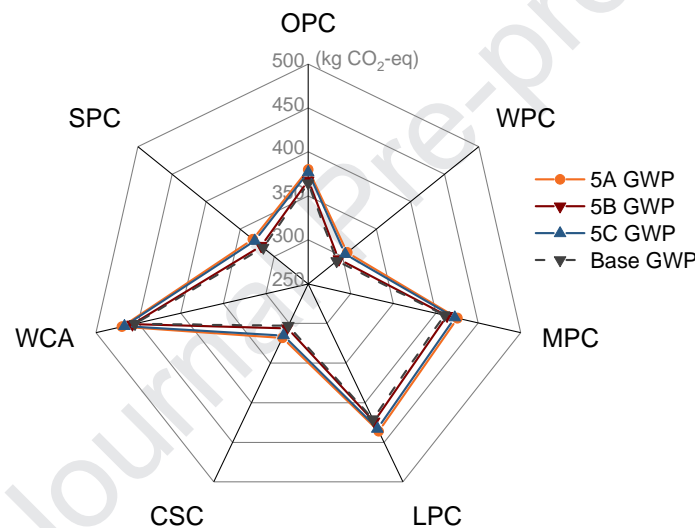


Fig. 8 Comparison of CO<sub>2</sub>-cured concrete blocks when changing transportation distance of raw materials

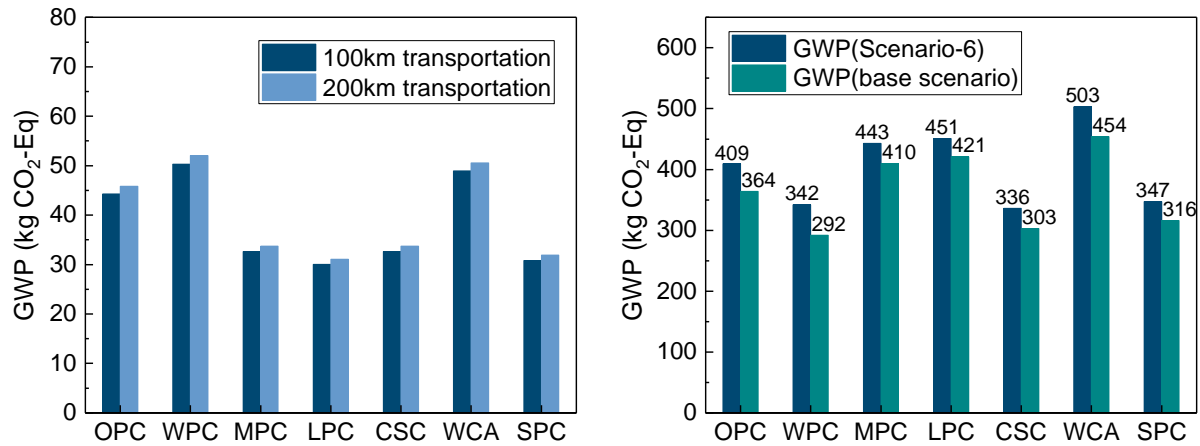


Fig. 9 (a) GWP increment for the CO<sub>2</sub> capture and cylinder transportation

(b) Total GWP in Scenario-6

## 4. Discussion and future perspectives

In terms of the different stages of material quarrying and manufacturing processes for CO<sub>2</sub>-cured concrete blocks, the reduced use of Portland cement should be regarded as a key step considering the primary GWP proportion accounted by Portland cement in cement-based binders (for Ordinary Portland Cement Block and other binary binders). Either the partial replacement of Portland cement or the use of Calcium silicate cement binder is beneficial to the avoidance of material GWP. Also using Portland cement leads to lower energy dependency than other binary binders, making it less effective to realize lower total emission and energy demand by saving energy consumption in manufacturing steps. When using binary binders, some carbonation-active minerals, naturally occurring wollastonite for instance, could help to achieve high CO<sub>2</sub> uptake by improving CO<sub>2</sub> internal diffusion (Wang et al., 2017).

Among these manufacturing processes, the energy consumption for maintaining high-pressure reaction accounts for the majority of total CED, leading to high values in terms of impact indicators. Its impact would become more significant with reduced curing duration. Thus, shortening the curing duration without compromising CO<sub>2</sub> uptake and compressive strength is of critical importance for effective environmental and energy consumption reduction.

Among the seven types of CO<sub>2</sub>-cured concrete blocks, the results presented demonstrate that the Wollastonite-Portland Cement Block and Slag-Portland Cement Block using natural wollastonite or BFS in binary binders are arguably the most sustainable. They not only

present the most favorably scores in all impact assessment indicators compared to the base case scenario, but also benefit significantly from the blending ratio increment and lightweight design. Using recycled waste aggregate in Waste Concrete Aggregate Block reduces the material GWP compared to Ordinary Portland Cement Block, but the energy consumption in the manufacturing process when applying long-time MC curing leads to high values of all impact indicators. Consider the construction waste issue around the world, the use of waste concrete aggregate for MC curing could show more benefit if its utilization is designed as a closed-loop process and the avoided burden of waste concrete aggregate is included. Similarly, high GWP from MC curing process also makes Limestone-Portland Cement Block less environmentally competitive, even though limestone reduces the GWP of Limestone-Portland Cement Block. The Calcium Silicate Cement Block corresponds to the least amount of material GWP, mostly because of the low calcination and reduced limestone use for producing calcium silicate cement (Sahu et al., 2013 ). Up to 16.8% reduction in total GWP could be realized when comparing Calcium Silicate Cement Block to Ordinary Portland Cement Block in the base scenario, and their other impact indicators are quite close. But the significant longer batch duration for MC curing of Calcium Silicate Cement Block should not be ignored, which may limit the manufacturing efficiency and increase investment. Further application of the cement-free Calcium Silicate Cement Block with MC curing would require a better understanding of the process optimization matching with rheological and hardening properties. Using the binary binder MgO-Portland Cement Block fails to realize effective GWP reduction in the base scenario. It should be mentioned that these results do not involve the consideration of mechanical improvement after MC curing. Until now, normalizing the impact indicators with respect to compressive strength or other mechanical indicators is unachievable, without the comprehensive experimental or industrial data among various CO<sub>2</sub>-cured products. Further investigation about the correlation of environmental impact indicators and strength performance is therefore required in the future.

In Fig. 10 the possible methods for the optimization of CO<sub>2</sub> mineral carbonation curing are presented. From the results of the scenario modelling, increasing the blending ratios in binary binders (except for MgO-Portland Cement Block) and the lightweight redesign prove to be strongly beneficial for reducing impact assessment indicators (GWP and CED) of CO<sub>2</sub>-cured concrete blocks, while the Wollastonite-Portland Cement Block and Slag-Portland Cement Block benefit the most. This suggests the potential benefit from material selection and optimization, especially the use of the low carbon binder with less energy consumption

for manufacture. Moreover, from a product density point of view, MC curing technology shows a better potential for emission reduction and energy saving when it is applied to the lightweight block product rather than the conventional block product. It was found that 1 m<sup>3</sup> CO<sub>2</sub>-cured lightweight concrete block could generate up to 56% avoidance benefit of carbon emission when compared to the conventional air-curing block with similar density. Also, the impacts of CO<sub>2</sub> uptake and energy consumption in manufacturing processes vary from different types of blocks. Wollastonite-Portland Cement Block and Ordinary Portland Cement Block show higher sensitivity to the variation of CO<sub>2</sub> uptake, while Limestone-Portland Cement Block, Waste Concrete Aggregate Block, and Calcium Silicate Cement Block present more significant variations in impact assessment indicators when changing the energy input of its manufacturing. Transportation of raw materials has limited impacts on indicators of GWP and CED within the studied distance (100 km), and results also indicated that the transportation distance of aggregate materials should be given the priority to that of binder materials. According to the results in Scenario-6, the current cost-ineffective capture and transportation of CO<sub>2</sub> would weaken the environmental competitive of CO<sub>2</sub>-cured concrete block if their impacts are included.

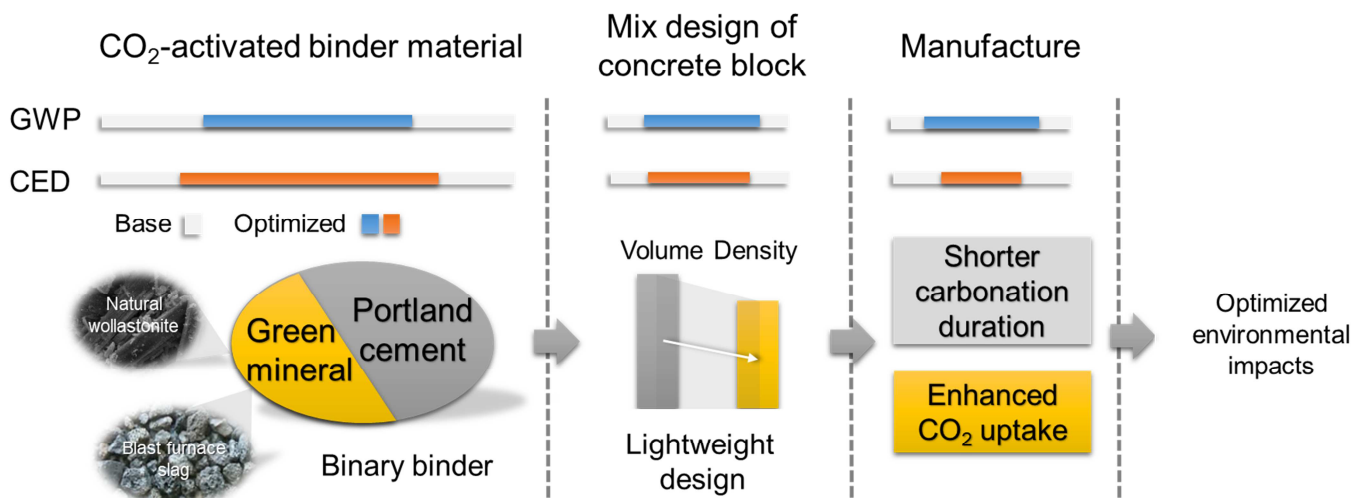


Fig. 10 Perspectives on the development of CO<sub>2</sub> mineral carbonation curing

## 5. Conclusions

This study conducted a comprehensive environmental LCA and evaluated the environmental impacts and influencing factors of seven promising CO<sub>2</sub>-cured concrete blocks from CO<sub>2</sub> MC curing processes. It was shown that 292~454 kg CO<sub>2</sub>-eq global warming potential (GWP) of 1 m<sup>3</sup> CO<sub>2</sub>-cured non-hollow concrete blocks were obtained, which is

1 lower than the 419kg CO<sub>2</sub>-eq GWP from the conventional steam-cured Ordinary-Portland  
2 cement block. From the point of view of materials and manufacturing, the reduced use of  
3 Portland cement is a key step for environmental optimization, while reducing the energy  
4 consumption for maintaining high-pressure carbonation helps to cut down the cumulative  
5 energy demand. Increasing the blending ratio in binary binders and the lightweight redesign  
6 also proved to be beneficial solutions for mitigating environmental impacts of CO<sub>2</sub>-cured  
7 concrete blocks. Transportation of raw materials has limited impacts on indicators of GWP  
8 and CED within the studied distance. Without the above optimization, the CO<sub>2</sub>-cured  
9 concrete block would show limited environmental advantage compared with steam cured  
10 products if the energy consumption and emission from the current CO<sub>2</sub> capture and  
11 transportation processes are included.



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### Nomenclature

$I_i$	Inventory value of the impact assessment indicator $i$	$IM_{CCT,i}$	Inventory value of indicator $i$ for energy input for CO <sub>2</sub> curing-Temperature (CCT)
$i$	Serial number of impact assessment indicator, equal to 1-12	$IM_{TP,i}$	Inventory value of indicator $i$ for material transportation
$IR_i$	Impact assessment data for raw material	$C$	Mass of CO <sub>2</sub> uptake of unit product (kg/m <sup>3</sup> )
$IM_i$	Impact assessment data for manufacturing processed	$D_j$	Transportation distance (equals to 50 km in the base scenario) of raw material $j$
$IT_i$	Impact assessment data for transportation of material	$IT_{j,i}$	Impact assessment indicator for lorry transportation (16-32 ton) (ton <sup>-1</sup> ·km <sup>-1</sup> )
$j$	Serial number of different raw material types	$Q_{CCT}$	Total heat needed for temperature maintenance in MC curing (kJ/m <sup>3</sup> )
$k$	Serial number of different production steps	$Q_R$	Heat needed to heat the CO <sub>2</sub> gas(kJ/kg)
$W_j$	Volume weight (kg/m <sup>3</sup> ) of raw material $j$	$Q_G$	Heat needed to heat the unit concrete block (kJ/m <sup>3</sup> )
$IM_{mix,i}$	Inventory value of indicator $i$ for mixing	$NG_{P,i}$	Inventory value of indicator $i$ from the production process to supply the corresponding heat
$IM_{comp,i}$	Inventory value of indicator $i$ for compaction	$NG_{U,i}$	Inventory value of indicator $i$ from the utilization (turbine) process of natural gas

$IM_{CCP,i}$	Inventory value of indicator $i$ for $t$ energy input for CO <sub>2</sub> curing-Pressure (CCP)	batch duration (h) which consisted of the duration of MC curing ( $t_C$ ) and the silence time ( $t_S$ ) between different batches
$T$	Stable temperature for MC curing (K)	$C_{p,G}$ Weight heat capacity of CO <sub>2</sub>
$W$	Total weight of unit concrete product (kg/m <sup>3</sup> )	$C_{p,R}$ Weight heat capacity of concrete block
$I_{MO,i}$	Inventory value of indicator $i$ of unit general machine operation time (m <sup>-3</sup> ·h <sup>-1</sup> )	

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**Acronyms**

GHG	Greenhouse gas	MC	Mineral carbonation
LCA	Life-cycle Assessment	LCI	Life cycle inventory
GWP	Global warming potential	CML-IA	Method for characterization developed by the Centre of Environmental Science in Leiden University
ISO	International Standards Organisation	OPC	Ordinary Portland Cement block
WPC	Wollastonite-Portland Cement Block	MPC	MgO-Portland Cement Block
LPC	Limestone-Portland Cement Block	SPC	Slag-Portland Cement Block
CSC	Calcium Silicate Cement Block	WCA	Waste Concrete Aggregate block
CCP	Reactor sealing, pressure control and gas circulation in MC curing	CCT	Reactor and gas heating, maintaining the temperature constant
CED-NRe	Cumulative energy demand of non-renewable energy resources	CED-Re	Cumulative energy demand of renewable energy resources
	Naturally occurring wollastonite	CCSC	Commercial calcium silicate cement
BFS	Blast furnace slag	AP	Acidification potential
EP	Eutrophication potential	HTP	Human toxic potential
POCP	Photochemical ozone create potential	FAETP	Freshwater aquatic ecotoxicity
ODP	Ozone layer depletion	MAETP	Marine aquatic ecotoxicity
ADP	Abiotic depletion	TETP	Terrestrial ecotoxicity

CED	Cumulative energy demand
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## References

- Ecoinvent, 2017. The Life Cycle Inventory Data Version 3.4. Swiss Centre for Life Cycle Inventories.
- Ahmad, S., 2003. Reinforcement corrosion in concrete structures, its monitoring and service life prediction-a review. *Cement & Concrete Composites* 25, 459-471.
- Ashraf, W., Olek, J., Jain, J., 2017. Microscopic features of non-hydraulic calcium silicate cement paste and mortar. *Cement & Concrete Research* 100, 361-372.
- Braga, A.M., Silvestre, J.D., Brito, J.D., 2017. Compared environmental and economic impact from cradle to gate of concrete with natural and recycled coarse aggregates. *Journal of Cleaner Production* 162, 529-543.
- Dahmen, J., Kim, J., Ouellet-Plamondon, C., 2017. Life cycle assessment of emergent masonry blocks. *Journal of Cleaner Production*, 171, 1622-1637.
- El-Hassan, H., Shao, Y., Ghouleh, Z., 2013. Effect of Initial Curing on Carbonation of Lightweight Concrete Masonry Units. *Aci Materials Journal* 110, 441-450.
- Faleschini, F., Zanini, M.A., Pellegrino, C., Pasinato, S., 2016. Sustainable management and supply of natural and recycled aggregates in a medium-size integrated plant. *Waste Management* 49, 146-155.
- Flower, D.J.M., Sanjayan, J.G., 2007. Green house gas emissions due to concrete manufacture. *International Journal of Life Cycle Assessment* 12, 282-288.
- García-Segura, T., Yepes, V., Alcalá, J., 2014. Life cycle greenhouse gas emissions of blended cement concrete including carbonation and durability. *International Journal of Life Cycle Assessment* 19, 3-12.
- Guinée, J.B., Gorée, M., Heijungs, R., Huppes, G., Kleijn, R., Koning, A.D., Oers, L.V., Sleeswijk, A.W., Suh, S., Haes, H.U.D., 2002. Handbook on LCA, operational guide to the ISO standards. ELSEVIER.
- Gursel, A.P., Masanet, E., Horvath, A., Stadel, A., 2014. Life-cycle inventory analysis of concrete production: A critical review. *Cement & Concrete Composites* 51, 38-48.
- Heede, P.V.D., Belie, N.D., 2012. Environmental impact and life cycle assessment (LCA) of traditional and 'green' concretes: Literature review and theoretical calculations. *Cement & Concrete Composites* 34, 431-442.
- Huang, H., Guo, R., Wang, T., Hu, X., Garcia, S., Fang, M., Luo, Z., Maroto-Valer, M.M., 2019. Carbonation curing for wollastonite-Portland cementitious materials: CO<sub>2</sub> sequestration potential and feasibility assessment. *Journal of Cleaner Production* 211, 830-841.
- ISO, 2006. Environmental Management - Life Cycle Assessment, Set of International Standards: ISO 14040-14044. International Organization for Standardization, Geneva.
- Liu, Q., Liu, J., Qi, L., 2016. Effects of temperature and carbonation curing on the mechanical properties of steel slag-cement binding materials. *Construction & Building Materials* 124, 999-1006.
- Mahoutian, M., Shao, Y., 2016. Production of cement-free construction blocks from industry wastes. *Journal of Cleaner Production* 137, 1339-1346.
- Marceau, M., Nisbet, M.A., Van Geem, M.G., 2007. Life cycle inventory of portland cement concrete. Portland Cement Association.
- Marinković, S., Dragaš, J., Ignjatović, I., Tošić, N., 2017. Environmental assessment of green concretes for structural use. *Journal of Cleaner Production* 154, 633-649.
- Mehta, P.K., Monteiro, P.J., 1993. Concrete: microstructure, properties, and material, second ed. Prentice-Hall, Upper Saddle River.
- Mo, L., Zhang, F., Deng, M., 2015. Effects of carbonation treatment on the properties of hydrated fly ash-MgO-Portland cement blends. *Construction & Building Materials* 96, 147-154.
- Mo, L., Zhang, F., Panesar, D.K., Deng, M., 2017. Development of low-carbon cementitious

- materials via carbonating Portland cement-fly ash-magnesia blends under various curing scenarios: a comparative study. *Journal of Cleaner Production*, 163, 252-261.
- Neville, A.M., 2000. *Properties of concrete*, fourth ed. Longman, London.
- Rostami, V., Shao, Y., Boyd, A.J., 2012. Carbonation Curing versus Steam Curing for Precast Concrete Production. *Journal of Materials in Civil Engineering* 24, 1221-1229.
- Ruan, S., Unluer, C., 2016. Comparative life cycle assessment of reactive MgO and Portland cement production. *Journal of Cleaner Production* 137, 258-273.
- Sahu, S., DeCristofaro, N., Cement™, S., 2013 Solidia Technology, White Paper. <http://solidiatech.com/> (accessed 13.03.16).
- Sanna, A., Uibu, M., Caramanna, G., Kuusik, R., Maroto-Valer, M.M., 2014. A review of mineral carbonation technologies to sequester CO<sub>2</sub>. *Chemical Society Reviews* 43, 8049-8080.
- Tu, Z., Guo, M.Z., Chi, S.P., Shi, C., 2016. Effects of limestone powder on CaCO<sub>3</sub> precipitation in CO<sub>2</sub> cured cement pastes. *Cement & Concrete Composites* 72, 9-16.
- Unluer, C., Al-Tabbaa, A., 2014. Enhancing the carbonation of MgO cement porous blocks through improved curing conditions. *Cement & Concrete Research* 59, 55-65.
- Vieira, D.R., Calmon, J.L., Coelho, F.Z., 2016. Life cycle assessment (LCA) applied to the manufacturing of common and ecological concrete: A review. *Construction & Building Materials* 124, 656-666.
- Wang, T., Huang, H., Hu, X., Fang, M., Luo, Z., Guo, R., 2017. Accelerated mineral carbonation curing of cement paste for CO<sub>2</sub> sequestration and enhanced properties of blended calcium silicate. *Chemical Engineering Journal* 323, 320-329.
- Yang, K.H., Jung, Y.B., Cho, M.S., Tae, S.H., 2015. Effect of supplementary cementitious materials on reduction of CO<sub>2</sub> emissions from concrete. *Journal of Cleaner Production* 103, 774-783.
- Yuan, F., Shen, L.Y., Li, Q.M., 2011. Emergy analysis of the recycling options for construction and demolition waste. *Waste Management* 31, 2503-2511.
- Zhan, B.J., Xuan, D.X., Chi, S.P., Shi, C.J., 2016. Effect of curing parameters on CO<sub>2</sub> curing of concrete blocks containing recycled aggregates. *Cement & Concrete Composites* 71, 122-130.
- Zhang, D., Ghoulah, Z., Shao, Y., 2017. Review on carbonation curing of cement-based materials. *Journal of CO<sub>2</sub> Utilization* 21, 119-131.